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## Polymeric Optical Films as Patterned Retarders and Alignment Layers for Transflective Liquid Crystal Displays

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*We developed a simple and versatile imprinting technique in combination with the exposure of ultraviolet light to fabricate several patterned optical wave plates using a polymerizable liquid crystalline (PLC) material. One of such patterned PLC films, produced by imprinting, can be used as both an in-cell retarder and an alignment layer for liquid crystal (LC) display applications. The chain ordering of the PLC molecules produced during imprinting results in anisotropic surface forces to align the LC molecules on the patterned PLC film without any surface treatment. A transflective LC display adopting an imprinted PLC film as an in-cell patterned retarder is demonstrated in a twisted nematic geometry.*

**Keywords:** imprinting technique; in-cell patterned retarder; liquid crystal (LC); polymerizable liquid crystalline (PLC) material; transflective LC display (LCD)

### 1. INTRODUCTION

Recently, polymeric functional materials have great interest due to their excellent performances in the optical and/or electronic devices as well as potential of molecular engineering for a new class of materials with appropriate physical and optical properties. Particularly, polymerizable liquid crystalline (PLC) materials have been widely used for producing brightness enhancement films [1], reflective polarizers [2,3], compensation films [4–6], and retarders [7–9] in liquid crystal displays (LCDs). In these cases, the PLC should be processed into an optically anisotropic film structure where an alignment layer is typically needed for orienting the PLC molecules.

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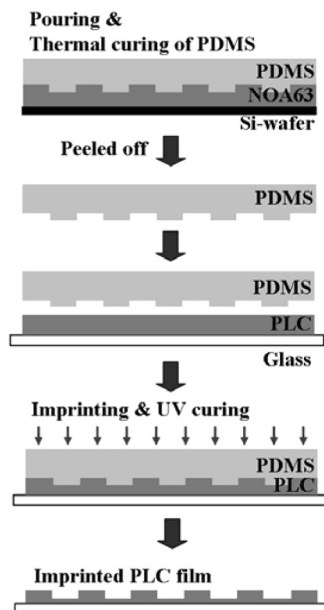
For producing the optically anisotropic PLC films, a rubbed surface [10] or photo-treated surfaces [11,12] have been used previously. A commonly used, rubbing process involves generation of electrostatic charges and dust particles from the mechanical contact. As an alternative to the rubbing process, a photoalignment technique has been extensively studied since it offers a unique way of preparing an alignment layer with complex structures for the anisotropic PLC films. However, the photoalignment suffers from the weak anchorage of the PLC on the photo-treated surface. Moreover, a baking process of an alignment layer at a high temperature over 120°C is required. Therefore, a new technique which is capable of aligning the PLC molecules at a low temperature needs to be developed for fabricating liquid crystal (LC) based microstructure devices.

In this work, we developed a simple and versatile imprinting technique, based on a soft-lithography [13,14], in combination with the exposure of ultraviolet (UV) light to produce a wide range of optically anisotropic films of the PLC material. For a certain LCD-type, transfective LCD consisting of transmissive and reflective subpixels [15], the use of an anisotropic PLC film, which serves as both an in-cell retarder and an alignment layer, is extremely important to achieve excellent optical performances and low cost production. The imprinting technique produces the chain ordering of the PLC molecules which results in anisotropic surface forces to align the LC molecules on the anisotropic PLC film without any surface treatment and extra alignment layers. The azimuthal anchoring energy was found to be on the order of  $10^{-5}\text{J/m}^2$ , which is sufficiently strong to produce stable electro-optic (EO) characteristics. In a twisted-nematic (TN) transfective LCD configuration, it was demonstrated that the imprinted PLC film behaves as an in-cell patterned retarder and induces two-domain alignment of the LC molecules.

## 2. OPTICALLY ANISOTROPIC PLC FILM

### 2.1. Fabrication of an Anisotropic PLC Film by Imprinting Technique

Figure 1 shows the schematic diagram of producing an optically anisotropic, patterned PLC film by the imprinting technique. The master made of a UV curable photopolymer material (NOA63, Norland Ltd.) on the Si-wafer was used to fabricate a polymer mold based on a poly(dimethylsiloxane) (PDMS, GE silicones) material. As shown as Figure 1, the PDMS was first poured on the master to give the



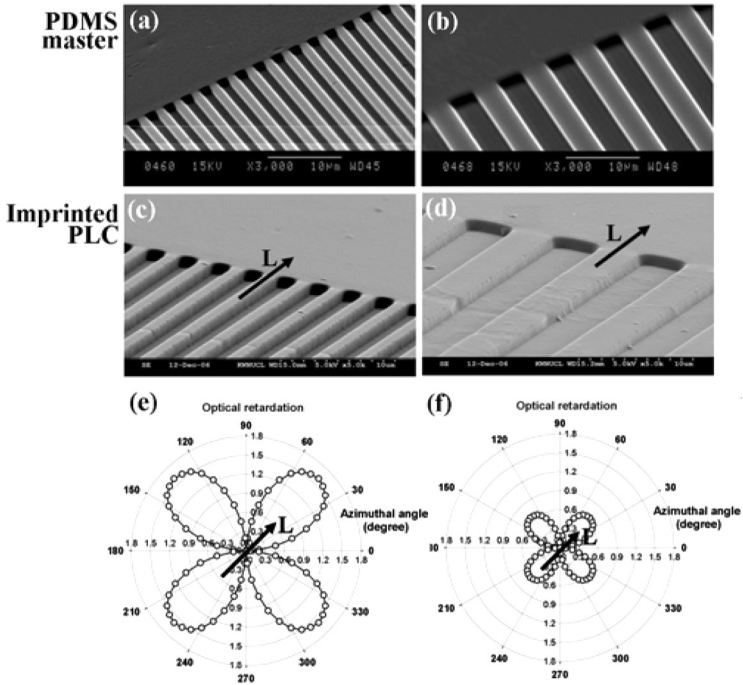
**FIGURE 1** The schematic diagram of producing a patterned PLC film by an imprinting technique.

polymer mold with thickness of 5 cm. It was then cured on a hot plate for 5 hours at about 70°C where the adhesion between the PDMS mold and the master was relatively small. The PDMS mold was finally peeled off from the master. The PDMS mold has micro-scale grooves shaped with the line to space (LS). The periods of the PDMS mold were 3.0  $\mu\text{m}$  and 8.0  $\mu\text{m}$  and the LS ratio of the mold was 1. The height of the micro-patterns of the PDMS mold was fixed as 1.2  $\mu\text{m}$ . The feature sizes were determined by using a scanning electron microscopy (SEM). We used a commercial PLC material, RMS 03-001 (E. Merck), to fabricate an optically anisotropic film. The PLC material was spin-coated on a indium-tin oxide (ITO) glass substrate at the spinning rate of 2500 rpm for 30 seconds without a pre-coated alignment layer, giving the layer thickness of 1.5  $\mu\text{m}$ , and baked subsequently at 65°C for 1 minute. By pressing the PDMS mold onto the prepared PLC layer, micro-scale LS grooves of the mold were replicated on the PLC layer. The micro-patterns of the PDMS mold were well transferred to the surface of the PLC layer under an imprinting pressure resulting from just above a gravitational force of the PDMS mold. After the micro-patterns were formed on the PLC layer, the PLC layer was exposed to the UV light at the intensity of 40 mW/cm<sup>2</sup> for

5 minutes under a nitrogen atmosphere so as to preserve the shapes of the micro-patterns. The optically anisotropic PLC film was then peeled off from the PDMS mold. The optical anisotropy was observed with a polarizing optical microscopy (POM, Optiphot2-Pol, Nikon) and the optical retardation was measured using a photo-elastic modulation (PEM) method [16].

2.2. Characteristics of the Imprinted Optically Anisotropic PLC Film

Figure 2 shows the SEM images of microgrooves produced on various PDMS molds and those transferred on the PLC films from the PDMS



**FIGURE 2** The SEM images of the PDMS molds and the imprinted PLC films, the negative line patterns of the PDMS molds, having different periods with same LS ratio from (a) to (d). The optical retardation of the patterned PLC films measured as a function of azimuthal angle by the PEM technique. The optical retardations of the patterned PLC films with different patterns are about 1.6 and 0.6 in (e) and (f), respectively. The different periods are  $3.0\mu\text{m}$  for (a), (c), and (e), and  $8.0\mu\text{m}$  for (b), (d), and (f). The black arrows (L) denote direction of the LS patterns in an imprinted PLC and the optical retardation results.

molds. The SEM images shown in Figures 2(a) and (b) correspond to the patterns having the periods of  $3.0\text{ }\mu\text{m}$  and  $8.0\text{ }\mu\text{m}$  with the LS ratio of 1. The height of the line patterns was about  $1.2\text{ }\mu\text{m}$ . Figures 2(c) and (d) show the SEM images of the imprinted PLC films, corresponding to the negative line shapes of the PDMS molds, whose periods are  $3.0\text{ }\mu\text{m}$  and  $8.0\text{ }\mu\text{m}$ , respectively. As shown in Figures 2(e) and (f), the optical retardation,  $2\pi d\Delta n/\lambda$ , of the optically anisotropic PLC film was measured using the PEM method. Here,  $d$ ,  $\Delta n$ , and  $\lambda$  represent the cell gap thickness, the optical anisotropy of the PLC film, and the wavelength of the light used, respectively. The measured results are shown in Figure 2(e) and (f). The maximum optical retardations of the optically anisotropic PLC film having the periods of  $3.0\text{ }\mu\text{m}$  with the LS ratio of 1 and  $8.0\text{ }\mu\text{m}$  with the LS ratio of 1 were about 1.6 and 0.6 in Figures 2(e) and (f), respectively. The directions of the maximum optical retardation of the imprinted PLC films agree well with that of the LS grooves of the films and the optical retardation values of the anisotropic PLC film having the period of  $3.0\text{ }\mu\text{m}$  correspond approximately to  $\lambda/4$  for  $\lambda = 632.8\text{ nm}$ . In general, it will also be possible to include the optically anisotropic PLC film in the LC cell to achieve a thinner and more durable display. Note that the PDMS mold has sufficient rigidity to prevent the collapse of microgrooves, and at the same time, it is soft enough to release the optically anisotropic PLC film. Using our imprinting process combined with the UV exposure, micro-scale lines will be replicated with high regularity at precisely determined micro-scale intervals, meaning that microgrooves can be replicated with high feature density and precision.

### 2.3. The LC Alignment on the Optically Anisotropic PLC Film

Through the imprinting process, an optically anisotropic PLC film was fabricated on the inner side of glass substrate. Using the Berreman concept [17], the microgrooves were prepared on the surface of the optically anisotropic PLC film to provide the spontaneous alignment of LC molecules on the PLC film without using any surface treatment or an extra alignment layer. The measured azimuthal anchoring energy, generated from the microgrooves, proved the possibility of aligning LC molecules by a TN LC cell implemented an imprinted PLC film. Our LC cell have an optically anisotropic PLC film, which optical retardation is same of that of a quarter-wave plate (QWP), on the lower glass substrate and a polyimide (PI) alignment layer of JALS 146-R50 (JSR Co., Japan) on the upper glass substrate. For fabricating TN LC alignment, the direction of micro-scale LS grooves of PLC film is perpendicularly aligned that of rubbing of the PI

alignment layer. The cell thickness ( $d$ ) was maintained  $5\text{ }\mu\text{m}$  thickness using glass spacers and the LC material used was MLC-6012 (E. Merck) and injected into the cell by capillary action at room temperature. The azimuthal anchoring energy was measured using the cell rotation method for the TN cell [18]. The azimuthal anchoring energy can be written as  $W_\varphi = 2K_{22}\varphi/d\sin 2\varphi$  where  $K_{22}$ ,  $d$ ,  $\varphi$ , and  $\varphi$  denote the twist elastic constant, the cell gap of the LC cell, and twist angle, respectively. Using the literature value of  $K_{22} = 5.5 \times 10^{-12}$ . In the microgrooves of the PLC films having the periods of  $3.0\text{ }\mu\text{m}$  with the LS ratio of 1, twist angle ( $\varphi$ ) and azimuthal anchoring energy ( $W_\varphi$ ) was  $84^\circ$  and  $1.55 \times 10^{-5}\text{ J/m}^2$ , respectively. The experimental values are in similar agreement with the theoretical prediction. It should be noted that the magnitude of the azimuthal anchoring energy generated by the microgrooves, on the order of  $10^{-5}\text{ J/m}^2$ , is sufficiently strong for aligning the LC over the film surface area from the theoretical point of view within the Berreman formalism [19]. On the other side, the measured azimuthal anchoring energy of the PLC films having the periods of  $8.0\text{ }\mu\text{m}$  was  $1.44 \times 10^{-6}\text{ J/m}^2$ , which is weak value for aligning the LC molecules.

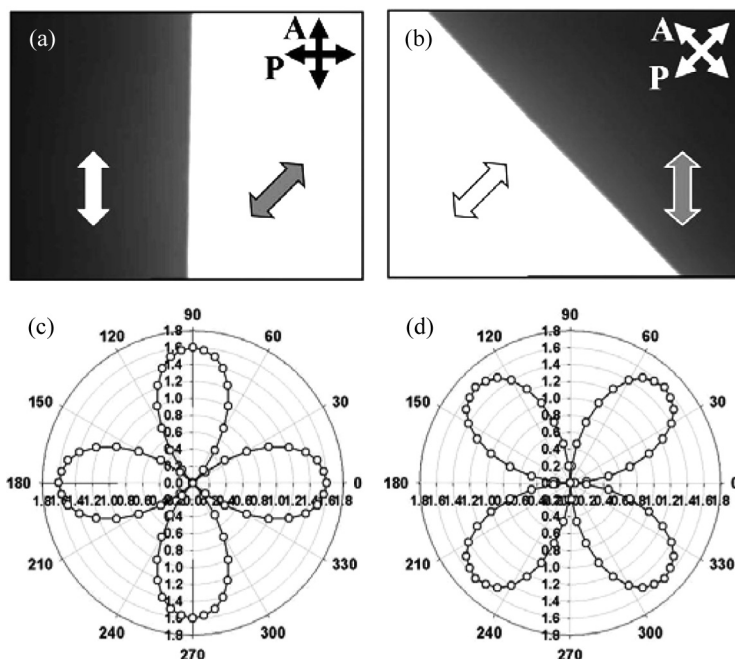
### 3. APPLICATIONS

#### 3.1. Optically Anisotropic, Patterned PLC Optical Films by Imprinting Technique

The optically anisotropic, patterned PLC films are very useful as polarization rotators [20] for stereoscopic devices, reflective color filters [11], polarization sensitive grating filters [21], and patterned optical retarders [22] in display applications. One example of an application of this type of layer in LCDs is to use these patterned PLC optical films for optical retardation wave plates. In particular, it is well known that QWP must be introduced in the LC cell for achieving reflective and transfective LCDs without loss of optical efficiency [23,24]. Using PDMS mold with LS patterns for imprinting process, this patterned optical retarder can be fabricated using an imprinting technique [25].

Figures 3(a) and (b) show microscopic textures of the optically anisotropic, patterned PLC film on the ITO glass substrate observed at an angle of  $45^\circ$  and  $0^\circ$  between the optic axis of the patterned PLC film and that of one of crossed polarizers using a POM. Two directions of LS in the patterned PLC give rise to different optic axes of the patterned PLC retarder. As shown in Figure 3(a), the patterned PLC retarder shows bright and dark states along the directions of  $45^\circ$





**FIGURE 3** The microscopic textures of bi-patterned PLC film on the ITO glass substrate without reflector observed under crossed polarizers. (a) an angle of  $0^\circ$  and (b)  $45^\circ$  between the direction of PLC film and the rear polarizer. Small white and gray arrows coincide with bi-domains with the direction of  $45^\circ$  and  $0^\circ$  aligned line patterns, respectively. The optical retardation of the bi-patterned PLC film measured as a function of azimuthal angle by the PEM technique. The optical retardations of the patterned PLC films with parallel direction and  $45^\circ$ -aligned direction respect to that of polarizer are about 1.6 in (c) and (d), respectively.

and  $0^\circ$  between the optic axis of the patterned PLC retarder as a QWP and the rear polarizer. Small white and gray arrows coincide with the directions of  $45^\circ$  and  $0^\circ$  aligned LS patterns, respectively. Figure 3(b) show microscopic textures when the patterned PLC retarder with two-domain was rotated by an angle of  $45^\circ$  with respect to Figure 3(a). In this case, the bright and dark states were reversed.

Figures 3(c) and (d) show the optical retardation of the patterned PLC retarder measured as a function of azimuthal angle by the PEM technique. The optical retardations of the patterned PLC retarders with parallel ( $0^\circ$ ) direction and  $45^\circ$ -aligned direction respect to that of polarizer are about 1.6, respectively as shown in

Figures 3(c) and (d). The measured optical retardations of the patterned PLC retarder in both two domains using the PEM method were all about  $\pi/2$  corresponds approximately to  $\lambda/4$  of the wavelength,  $\lambda = 632.8 \text{ nm}$ , used. But, the directions of the maximum retardation in the  $0^\circ$ -aligned and the  $45^\circ$ -aligned direction respect to that of polarizer were  $0^\circ$  and  $45^\circ$ , respectively. Thus, the results are same as the directions of the LS patterns in two domains of the imprinted PLC film.

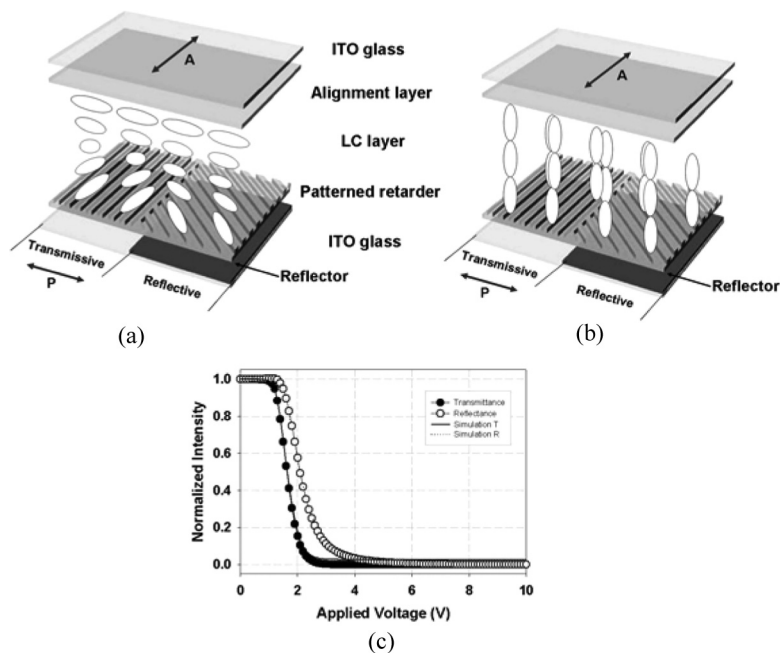
### 3.2. Transflective LCD With an In-cell Patterned Retarder

In the ubiquitous environment, the patterned PLC retarders [26,27] are essential to produce transflective LCDs for mobile applications. The in-cell patterned retarders described here have several advantages such as the compactness, light weight, and no parallax over an external optical film when fabricated directly onto the interior of the LCD. Furthermore, due to develop the PLC-based optical components by imprinting technique, a novel transflective LC cell without alignment layers in a simple and cheap manner was obtained.

Figure 4 show the structure, operation principles and EO properties of our transflective LC cell with multimode configuration consisting of the  $90^\circ$ -TN mode in the transmissive region and the  $45^\circ$ -TN mode in the reflective region.

The microstructures were prepared on the PLC film to explore the possibility of aligning the LC molecules without using an extra alignment layer and/or any surface treatment. An optically anisotropic, patterned PLC film was implemented into a LC cell to examine two-fold functionality of an alignment layer and an in-cell patterned optical retarder. An array of aluminium (Al) reflectors with the periodicity of  $100 \mu\text{m}$  was prepared on the ITO glass substrate. As shown in Figures 4(a) and (b), our transflective LC cell have an imprinted, patterned PLC retarder on the lower ITO glass substrate with an array of Al reflectors and a PI alignment layer of JALS 146-R50 (JSR co., Japan) on the upper glass substrate. For the TN LC alignment, direction of rubbing on the PI alignment layer is perpendicular to that of  $0^\circ$ -aligned domain of the PLC retarder. The LC cell was placed between crossed polarizers such that two LC modes,  $45^\circ$ -TN state in the transmissive region and a  $90^\circ$ -TN state in the reflective region, due to directions of the microstructures on the surface of the imprinted PLC film.

The nematic LC material used in our transflective LC cell was MLC-6012 (E. Merck). The material parameters used for numerical simulations were the elastic constants,  $K_1 = 11.6 \times 10^{-12} \text{ N}$ ,  $K_2 = 5.5 \times 10^{-12} \text{ N}$ ,  $K_3 = 16.1 \times 10^{-12} \text{ N}$ , the ordinary refractive index



**FIGURE 4** The structure and operation principles of our transflective LC cell with bi-domains configuration consisting of the 90°-TN mode in the transmissive region and the 45°-TN mode in the reflective region (a) a bright state (field-off) and (b) a dark state (field-on). The EO characteristics of our transflective LC cell. The open circles, the filled circles, solid lines, and dotted lines denote the experimental results of the transmittance, the reflectance, numerical simulations of the transmittance and the reflectance, respectively.

$n_o = 1.4620 + 5682/\lambda^2$ , the extraordinary refractive index  $n_e = 1.5525 + 9523/\lambda^2$ , the dielectric anisotropy  $\Delta\epsilon = 8.2$ , and the rotational viscosity  $\gamma_1 = 0.192$  Pa · sec. Here,  $\lambda$  is the wavelength of the incident light in nanometer. The cell thickness was maintained  $4\mu\text{m}$  thick using glass spacers so that the LC material injected into the cell by capillary action at room temperature. Figure 4(c) shows the experimental EO results and those of numerical simulations for our transflective LC cell as function of the applied voltages. The transmittance and the reflectance were normalized to examine the essential features of the EO response in both the transmissive and the reflective regions. The symbols and the lines denote the experimental results and the numerical simulations of the transmittance and the reflectance, respectively. The experimental EO results agree well simulation results.

## 4. CONCLUSION

We developed an anisotropic imprinting technique, combined with the exposure of UV light, to produce a patterned optical film of a PLC material. The patterned PLC film can be used as both an in-cell optical retarder and an alignment layer of the LC molecules for the LC-based optical devices including transfective LCDs. The magnitude of the orientational order of the PLC molecules, acquired during the imprinting process at a micro-scale level, depends on the feature size, the adhesion, and the surface properties of the PDMS mold. The imprinting technique presented here provides a fast, low-cost process for the parallel replication of two-dimensional anisotropic microstructures of a PLC material.

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